Nested Fermi Surface Segments, a Pseudogap, and Nanoscale Fluctuating Charge/Orbital Ordering in Colossal Magnetoresistive (CMR) Oxides

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ABSTRACT

Using angle-resolved photoemission, we have made the first direct measurements of the key transport parameters of the metallic state of $La_{1.2}$ $Sr_{1.8}$ Mn_2 O_7, including the full 2d Fermi surface, Fermi velocity, effective mass, mean free path, and mean free time between scattering events. Using these parameters we calculate the in-plane Drude conductivity, which turns out to be roughly an order of magnitude higher than the measured DC conductivity. Absent from the Drude calculation is the pseudogap, which we find to remove at least 90 percent of the spectral weight at E_F, consistent with this disagreement. The Fermi surface is dominated by parallel straight sections making it highly susceptible to nesting instabilities. The Q vectors that nest this Fermi surface are observed with short range order in diffraction experiments, making the presence of nanoscale fluctuating charge/orbital density wave gaps the likely origin of the pseudogap and a critical component of CMR physics.

EXPERIMENTAL

We used the angle mode of the High Energy Resolution Spectrometer (HERS) at beamline 10.0.1 to simultaneously measure spectra over a ±8 degree angular space with an angular resolution of better than ± .16 degrees and an energy resolution of about 15 meV. We cleaved single crystalline samples of the layered manganite $La_{1.2}Sr_{1.8}Mn_2O_7$ which had a T_c of 126K. The data shown here were cleaved and measured at a temperature of 20K at a vacuum of $4x10^{-11}$ torr.

RESULTS

We studied the low temperature ferromagnetic metallic state of cleaved single crystals of $La_{1.2}$ $Sr_{1.8}$ Mn_2 O_7 for these studies. Figure 1 presents the near E_F spectral weight measured throughout the Brillouin zone, giving an experimental mapping of the two-dimensional Fermi surface. We overlay this intensity plot with the Fermi surface topology predicted by a Spin-polarized Local Density Approximation (LSDA) calculation (black lines)[1,2]. This theoretical Fermi surface consists of mainly two parts: one small electron pocket centered around the zone center (0,0) which has predominantly out-of-plane $d_{3z^2-r^2}$ character, and two concentric large hole pockets centered around the zone corners (±π,±π) which have predominantly in-plane $d_{x^2-y^2}$ character. It is seen that the experimental image plot matches the theoretical Fermi surface relatively well: the higher intensity around (0,0) corresponds to the small electron pocket and four L-shape loci around the corners correspond to portions of the hole pockets.

More details of the electronic structure can be obtained through the study of E versus k dispersion relations, which we have directly measured in these experiments. The standard EDCs (Energy distribution Curves) are very broad and asymmetric and the dispersion cannot be followed to the Fermi energy using them. However, by analyzing the data in terms of Momentum Distribution Curves or MDCs, much more information can be extracted because the MDCs have simple Lorentzian lineshapes which can be easily fit. Figure 2a shows a stack of MDCs cutting

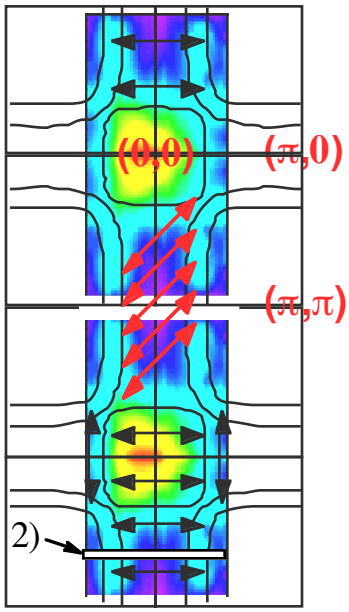

Fig 1. Experimental Fermi Surface of of $La_{1.2}$ $Sr_{1.8}$ Mn_2 O_7. Curved lines are calculated FS's and red and black arrows are superlattice vectors from X-ray scattering [7].

through the in-plane FS and figures 2b and 2c show the fitting results, which enable us to extract the wave vector k, the width Δk and the spectral weight of the peaks for each MDC, which can then be plotted against binding energy. This gives the key transport parameters for these in-plane states, which we find to be essentially independent of location along the roughly one-dimensional Fermi surface. The Fermi velocity v_F determined by the slope of the dispersion at E_F, $v_F = (1/\hbar)(dE/dk\}$, is ~ 0.038c. The effective mass m* from fitting the experimental dispersion relation with a parabola is ~ 0.27 m_e, which is within ~10% of the band theory result for these states. This very light mass is surprising for these compounds, particularly in light of the widespread discussion of polaronic coupling in these materials [3] which is believed to extend into the low temperature state as well as the high temperature state.

The angular width at E_F is around 1.25° HWHM, or equivalently the momentum width is $0.09\pi/a \sim 0.07$ Å^{-1}. Since our angular resolution is significantly better than this, we consider it to be intrinsic and originating from the finite electron mean free path $\lambda=1/\Delta k \sim$ 14 Å. This mean free path is roughly 7 times the in-plane spacing between the Mn and O atoms. A similar value has been estimated by Gray et al from the magnetic-field dependence of the in-plane conductivity[4], which they fit to the theoretical form of quantum interference effects suggested by Abrikosov [5]. We also can calculate the mean free time between scattering events $\tau=\lambda/v_F$, obtaining a value ~ 1.24 fs. Finally, we can estimate the number of in-plane carriers from the volume of the Fermi surface pockets centered around (π,π), which cover nearly 1/2 of the full Brillioun zone size, or n ~ $3.4*10^{21}$ holes/cm^3.

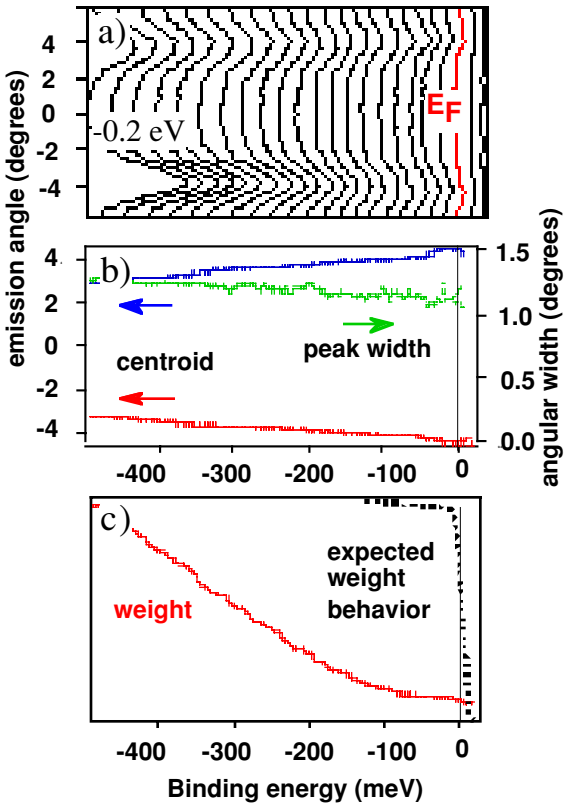

Fig 2 a) MDC's along the cut labeled 2) in figure 1. b) Centroids and widths from Lorentzian fits of the MDCs .c) Weight of the MDCs and expected weight behavior

From all these parameters the in-plane DC conductivity can be estimated based on the Drude formula: $\sigma=ne^2\tau/m^*$, where we note that all the parameters are approximately independent of the location on the in-plane Fermi surface (the Fermi surface centered around (0,0) will of course have very different parameters, but these will not be very relevant for the in-plane conductivity). Plugging in the numbers we obtain the resistivity $\rho_{ARPES}=1/\sigma \sim 2*10^{-4}$ Ω-cm while the measured low-temperature resisivity $\rho_0 \sim 2*10^{-3}$ Ω-cm is nearly one order of magnitude larger than ρ_{ARPES}. The point is that the low temperature state of these materials is a very poor metal with the resistivity on the verge of the inverse of Mott's minimum metallic conductivity. Such poor conductivity would typically be regarded as the outcome of either very strong scattering (a short mean free path on the order of the lattice constant) or a very small number of carriers (small Fermi surface), although our data directly shows that neither can be the case. It may also be considered to originate from polaronic effects, although we do not observe the large mass enhancement that would typically associate with it.

In order to resolve this inconsistency other physics must be included, the most obvious of which is the suppression of spectral weight near the Fermi energy. Figure 2(b) shows the very unusual behavior that the spectral weight of the dispersive peak falls rapidly over quite a large energy scale (nearly half an eV) as the peak approaches E_F. This behavior was previously termed a ``pseudogap". It is not expected in the simple theory of metals, in which the spectral weight remains constant until it is cut by the Fermi function (black dotted line; this is exactly true for one-dimensional systems

with a linear dispersion relation, which is closely approximated here). This pseudogap will effectively decrease the conductivity by removing a large portion of the carriers from the conduction process. As the temperature is raised above T_c, our previous work showed that the pseudogap completely removes all remaining spectral weight at E_F, explaining the insulating behavior of the high temperature phase[6]. Therefore, the pseudogap appears critical for explaining the poor conductivity of both phases, and presumably should be important for explaining the transition between them as well.

Looking back at figure 1, we notice that the measured Fermi surface segments around the zone corners are somewhat straighter than predicted by the calculation, indicating a propensity for nesting instabilities. X-ray scattering experiments have recently been carried out on these materials [7] and two sets of weak superlattice diffraction peaks not corresponding to the regular crystal structure have been uncovered. The Q-vectors of these superlattice peaks is shown in the figure and these are shown to almost nest (red lines, CE ordering vector with Q=0.25,0.25,0) and perfectly nest (black lines, "orbital stripes" vector with Q=0.3,0,1) the measured FS. Therefore, both orderings may affect (gap) the near E_F electronic states. The agreement also gives a natural explanation to the origin of the orbital stripes superlattice peaks - they should arise as a result of the FS nesting between nearly straight Fermi surface segments, with the nesting feeding back to enhance the straightness of the FS segments. We thus argue that we should consider the absence of near E_F spectral weight above T_c as originating primarily from a charge-density wave style gap due to the (0.3,0,1) ordering as well as to a degree the CE ordering. The fact that essentially the entire FS can be affected by these vectors (see figure 1) means that there will be no un-gapped portions remaining, consistent with the insulating behavior above T_c. At higher temperature where the gap is stronger and more static, our preliminary measurements do show the bending back behavior of the bands expected from a CDW or SDW style gap, although this is very weak both because of the coherence factors and fluctuating nature of the orderings.

The data from reference [6] as well as that from optical experiments [8] show that the pseudogap begins to fill in as the temperature is lowered through T_c. This is consistent with the weakening of the orderings in the metallic phase. Although the scattering experiments cannot observe them in the ferromagnetic phase [7] a pseudogap is still observed (to a lesser degree) and the Fermi surface segments are still very straight. We take this as the indication for the nanoscale remnants of the orderings below T_c: they fluctuate violently in both space and time such that the scattering experiments are not able to observe them, but they nonetheless still very much affect the electronic structure. Of course, the fluctuations give the gap many energy scales and make the gap edges rather ``soft" in energy. In this picture, the insulator-metal transition and hence CMR will occur just when the charge/orbital orderings are strong enough to remove all the weight at E_F.

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